Electronic Structure Calculations for $YBa_2Cu_3O_7$ within the Slave Boson Formalism

M. Biagini

Istituto Nazionale per la Fisica della Materia and Dipartimento di Fisica, Universitá di Modena, via Campi 213/A, I-41100 Modena, Italy

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A novel method for self-consistent electronic structure calculations for correlated metals is presented. This approach takes into account on-site Coulomb repulsion among localized electrons and is based on the slave boson formalism. The proposed method was applied to the study of the electronic structure of $YBa_2Cu_3O_7$: the on-site Coulomb repulsion of Cu $3d$ electrons gives origin to a flat band at the Fermi level, as observed in experiments. A sharp Van Hove singularity has been found 25 meV below the Fermi energy. [S0031-9007(96)01615-8]

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The density functional theory (DFT) [1] has proved very successful in describing ground state properties of several physical systems. The local density approximation (LDA) and the local spin density approximation (LSDA) have provided a good description of some of the properties of high T_C superconductors, such as, e.g., the lattice dynamics and crystal stability [2]. Nevertheless, such approximations fail to predict the insulating, antiferromagnetic ground state of the parent undoped materials. Similar problems were observed much earlier in transition metal oxides such as FeO, CoO, and NiO. Such a failure is due to the fact that LSDA does not treat adequately the many-body Coulomb interaction in materials which exhibit a strong electron-electron interaction. Several attempts to improve LSDA have been made. Both self-interaction correction (SIC) [3] and local spin density approximation with on-site Coulomb correlation (LSDA + U) [4] have been successfully applied to the study of undoped cuprates [5].

In these materials, the state with the highest energy has mainly a Cu $3d_{x^2-y^2}$ character, which is singly occupied in undoped compounds. The insulating character is due to the on-site Coulomb repulsion, which opposes transfer processes involving a doubly occupied Cu $3d_{x^2-y^2}$ orbital. In doped cuprates, charge transfer processes become possible, so that such systems show a metallic behavior. Nevertheless, correlation effects are still present in these systems, which exhibit anomalous normal-state properties and short-range antiferromagnetic correlations.

Recently, angle-resolved photoemission spectroscopy (ARPES) experiments have shown evidence of interesting features in the electronic structure of high-T_C superconductors [6]. In particular, these experiments have shown the presence of an extended saddle point near the Fermi level. Several hypothesis have been made about the role that a van Hove singularity close to the Fermi level could have in determining the physical properties of both normal and superconducting states [7]. The nature of the flat-band saddle points is still controversial. Andersen *et al.* [8] have attributed them to band structure effects, related to the dimple of the CuO_2 planes. On the other hand, many authors believe that such features are originated by many-body effects, and cannot be explained within a single-particle framework. Recently, several groups [9] have presented results obtained for the Hubbard and t-J models, which show flat bands at the Fermi level, similar to those observed in the experiments.

From the above considerations, the need of an adequate theory for electronic structure calculations for the metallic state of cuprates results is evident. In the present Letter, I describe a novel method for self-consistent electronic structure calculations for correlated metals. Such a method has been applied to the study of the electronic structure of $YBa_2Cu_3O_7$ (YBCO7).

In the strong-coupling limit, when the Coulomb repulsion on the Cu sites is supposed much larger than the bandwidth, the Hamiltonian of the system can be set under the constraint that no Cu $3d_{x^2-y^2}$ orbital is doubly occupied. Such a constraint can be satisfied by employing the slave boson formalism [10], in which the electron operator $d^\dagger_{i\sigma}$, corresponding to the Cu $3d_{x^2-y^2}$ state, is replaced by $d^\dagger_{i\sigma} \rightarrow f^\dagger_{i\sigma} b_i$. The slave boson technique has been applied to the study of several mean-field phases of the t-J model [11]. The boson operator b_i keeps track of the empty sites, and the fermion operator $f_{i\sigma}$ carries the spin. The constraint of no double occupancy is satisfied by requiring that $b^\dagger_i b_i + \sum_\sigma f^\dagger_{i\sigma} f_{i\sigma} = 1$ at each site i; this request may be reached by adding to the Hamiltonian a term

$$\sum_i \lambda_i \left(\sum_\sigma f^\dagger_{i\sigma} f_{i\sigma} + b^\dagger_i b_i - 1 \right), \qquad (1)$$

where λ_i is a Lagrangian multiplier. A mean-field theory is achieved by replacing the fields b_i and λ_i by their saddle-point values b_0 and λ_0. The mean field λ_0 can be interpreted as the chemical potential μ_b of bosons.

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The basis set used in this work is the one corresponding to the linear-muffin-tin orbital method, in the atomic sphere approximation (LMTO-ASA) [12,13]. It must be stressed that the LMTO orbitals do not provide an exactly orthogonal set. Nevertheless, the $3d$ orbitals are well localized and can be easily associated with a given site. The LMTO-ASA method has been used in several high-T_c^0 superconductor band structure calculations [14–16] and is found to give very similar results to those obtained through more elaborate methods such as the full-potential linear augmented plane-wave method (FLAPW) [17]. I have performed self-consistent electronic structure calculations for YBCO7. In these calculations, all electrons were separated into either core or band electrons. The core electron wave functions were calculated by solving the relativistic Dirac equation at every self-consistent iteration, while the spin-orbit coupling was neglected for the valence electrons. All calculations were performed for the orthorhombic structure with lattice constants and atomic position parameters taken from Ref. [18]. An empty sphere was put along the x axis as described in Refs. [14,15]; 147 k points within the irreducible wedge of the Brillouin zone were used in the self-consistent calculations.

In order to obtain the mean-field slave boson Hamiltonian H^{SB}, corresponding to the LMTO Hamiltonian H [see Eq. (6.17) in Ref. [13]], it is useful to consider the following rules: A local term $d_{i\sigma}^{\dagger} d_{i\sigma}$ is not modified and becomes $f_{i\sigma}^{\dagger} f_{i\sigma}$; this result can be obtained by using the relations $b_i b_i^{\dagger} = b_i^{\dagger} b_i + 1 = 2 - \sum_{\sigma} f_{i\sigma}^{\dagger} f_{i\sigma}$ and $f_{i\sigma}^{\dagger} f_{i,-\sigma} = f_{i\sigma} f_{i,-\sigma} = 0$, which follow from the constraint of no double occupancy. A hopping term $t_{d\alpha} \sum_{\sigma} a_{\alpha i\sigma}^{\dagger} d_{j\sigma}$ between a planar Cu(2) $3d_{x^2-y^2}$ state and a different state $|\alpha\rangle$ is replaced by $b_0 t_{d\alpha} \sum_{\sigma} a_{\alpha i\sigma}^{\dagger} f_{j\sigma}$. Hopping terms which do not involve the planar Cu(2) $3d_{x^2-y^2}$ orbitals remain unchanged. The Hamiltonian assumes then the following form (for simplicity it is considered the case of a single atom per cell)

$$\underline{\underline{H}}^{\mathrm{SB}} = \underline{\underline{b}}\,\underline{\underline{H}}\,\underline{\underline{b}} + \underline{\underline{\gamma}}, \tag{2}$$

where the matrix $\underline{\underline{b}}_{ij}$ is given by

$$\underline{\underline{b}}_{i,j} = \begin{pmatrix} \frac{b_0+1}{2} & \frac{b_0-1}{2} & 0 & 0 & \cdots & 0 \\ \frac{b_0-1}{2} & \frac{b_0+1}{2} & 0 & 0 & \cdots & 0 \\ 0 & 0 & 1 & 0 & \cdots & 0 \\ \cdot & \cdot & \cdot & \cdot & \cdots & \cdot \\ 0 & 0 & 0 & 0 & \cdots & 1 \end{pmatrix}, \tag{3}$$

where the index $i, j = 1$ corresponds to $(l = 2, m = -2)$ and $i, j = 2$ to $(l = 2, m = 2)$. The matrix $\underline{\underline{b}}_{ij}$ has been introduced to modify the hopping term between a Cu(2) $3d_{x^2-y^2}$ state and another state, and between two Cu(2) $3d_{x^2-y^2}$ orbitals in different sites. The matrix $\underline{\underline{\gamma}}$ is introduced to correct the local terms, which must remain unchanged, and to take into account the constraint given in Eq. (1). The matrix $\underline{\underline{\gamma}}$ is given by

$$\underline{\underline{\gamma}}_{i,j} = \begin{cases} \dfrac{N\lambda_0}{2} - \dfrac{M(b_0^2 - 1)}{2}, & \text{if } i = 1,2 \text{ and } j = 1,2, \\ 0, & \text{elsewhere}, \end{cases} \tag{4}$$

where N is the normalization constant for the muffin-tin orbital Cu(2) $3d_{x^2-y^2}$ and

$$M = \left[\frac{2\omega(-) + 2E_{\nu}[1 + \omega^2(-)\langle \dot{\phi}^2 \rangle]}{S\phi(-)^2} \right]_{l=2} + \sum_{l'} D_{l'} \left[\frac{2\omega(+) + 2E_{\nu}[1 + \omega^2(+)\langle \dot{\phi}^2 \rangle]}{S\phi(+)^2} \right]_{l'}. \tag{5}$$

The coefficient $D_{l'}$ is given by

$$D_{l'} = \sum_{m'=-l'}^{l'} \sum_{m=\pm 2} \sum_{\mathbf{R}\neq 0} (g_{l',m',l=2,m})^2 \left(\frac{S}{r}\right)^{2l'+5} \times |Y_{l'+2,m'-m}(\hat{\mathbf{R}})|^2 \frac{\pi}{(2l'+1)^2}, \tag{6}$$

where $\mathbf{R}$ is a lattice vector, and $g_{l',m',l,m}$ are defined in Eq. (6.8) in Ref. [13].

The mean field b_0 is related to the average value of doping holes on the planar Cu sites. Holes introduced by doping have predominantly O $2p$ character, because of the strong Coulomb repulsion among Cu $3d$ states [19]. In the case of YBCO7, experimentally the doping hole content is ≈ 0.2 holes per CuO_2 plane, and assuming [20] that such holes have 15% Cu $3d$ character, one obtains the value $b_0^2 = 0.03$. This choice corresponds to the calculations presented in this work; the use of different values of b_0 (e.g., $b_0^2 = 0.01$ or $b_0^2 = 0.1$) do not modify significantly the electronic structure. In principle, the Lagrangian parameter λ_0 should be determined through the minimization of the total energy of the system with respect to λ_0. This procedure would assure one that the occupancy of the Cu(2) $3d_{x^2-y^2}$ orbital, given by $\langle \sum_{\sigma} f_{\sigma}^{\dagger} f_{\sigma} \rangle$, is equal to $1 - b_0^2$. Unfortunately, the calculated value of the above occupancy is not in good agreement with the experimental one. Therefore, the value of λ_0 has been determined by imposing the alternative constraint $\sum_{\sigma} \langle f_{\sigma}^{\dagger} f_{\sigma} \rangle = n_0^{\mathrm{LDA}}$, where n_0^{LDA} is the occupancy of the orbital as obtained in the ordinary LDA calculations. This procedure would make the LDA and LDA + SB results comparable (LDA + SB is the acronym I will use to refer to the method presented herein).

In Fig. 1 the LDA + SB (solid lines) total and partial densities of states of YBCO7 are compared with the corresponding LDA results (dashed lines). The notations Cu(2), O(2), and O(3) refer to the sites in the CuO_2 planes, while Cu(1) and O(1) to those in the CuO chain. A sharp peak, located 25 meV below the Fermi level, has been found in the partial density of states corresponding to the Cu(2) sites. This feature is not present in LDA results,

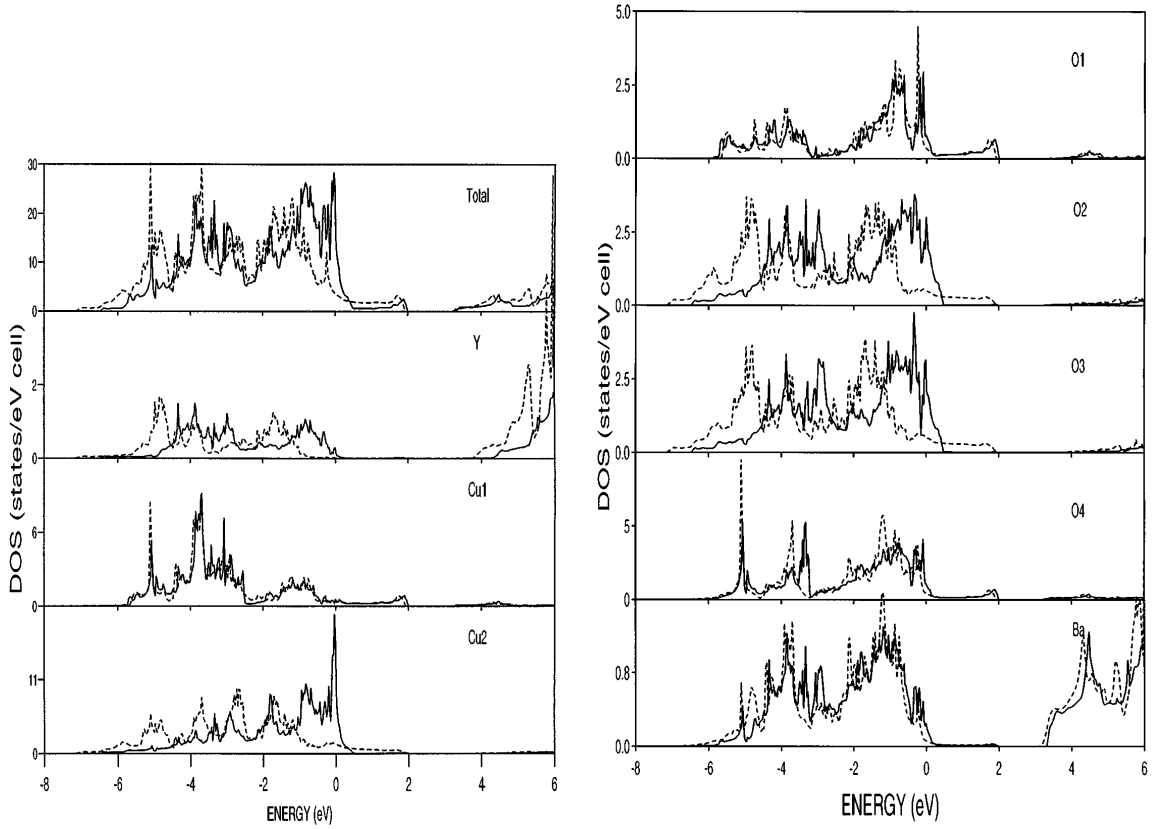

FIG. 1. Total and partial densities of states of YBCO7, as obtained from LDA + SB (solid lines) and LDA (dashed lines) self-consistent electronic structure calculations. The energies are referred to the Fermi level.

and indicates the existence of a van Hove singularity close to the Fermi energy. The electronic structure of the CuO_2 planes is strongly modified, with respect to the LDA results, while only minor changes have been found in the densities of states corresponding to the CuO chains. The study (not shown here) of the projections of the local partial DOS for Cu(2) $3d$ on irreducible representations of the point group D_{4h} has shown that the van Hove singularity at the Fermi level is present only in the Cu $3d_{x^2-y^2}$ component; nevertheless, significant changes have been observed in the remaining Cu $3d$ states, which are placed closer to the Fermi level with respect to the LDA results.

The LDA + SB band structure of YBCO7, represented in Fig. 2, differs significantly from the LDA band structure. The most striking feature is the existence of a flat band at $E_F \pm 25$ meV along the direction Γ-X, which has a mixed Cu(2) $3d$-O$(2,3)$ $2p$ character. The results presented in this work suggest that the extended saddle points observed in experiments are originated by electron-electron correlation in the planar Cu(2) sites. This conclusion is in agreement with those reached in recent studies of the Hubbard and t-J models [9]. On the other hand, this view contrast with the one presented by Andersen *et al.* [8]; these authors do not consider correlation effects and identify the O$(2,3)$ $2p_z$ orbitals as the relevant electronic degrees of freedom responsible for the existence of a flat band at the Fermi level.

In conclusion, I have presented a novel method for self-consistent electronic structure calculations for strongly correlated metals. The present method is developed within the slave boson formalism, and allows one to take into account on-site Coulomb repulsion among localized

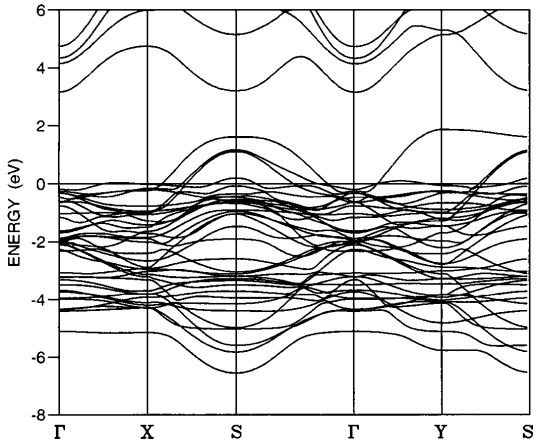

FIG. 2. Band structure of YBCO7, as obtained from LDA + SB self-consistent electronic structure calculations. The energies are referred to the Fermi level.

states. This approach is suitable to the study of the electronic structure of cuprates at optimal doping, because of the strong correlation of Cu 3*d* electrons. Self-consistent electronic structure calculations have been performed for YBCO7, which shows that on-site Coulomb repulsion is responsible for the existence of a flat band at the Fermi level, as observed in experiments. The density of states presents a novel feature with respect to LDA results, which consists of a sharp peak positioned 25 meV below the Fermi energy.

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